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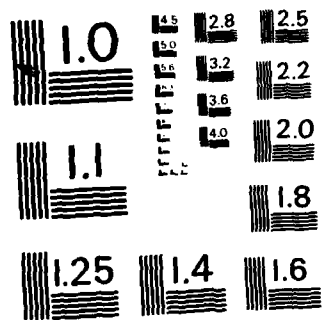
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**CHEMICAL
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US Army Armament Research and Development Command
Aberdeen Proving Ground, Maryland 21010

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TECHNICAL REPORT

ARCSL-TR-83006

**PREDICTING THE YIELD PERFORMANCES OF MULTICOMPONENT
OIL SMOKES CONFINED IN CLOSED CHAMBERS**

by

Glenn O. Rubel

Research Division

June 1983



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| 20. ABSTRACT (Continue on reverse side if necessary and identify by block number) A semiempirical model is developed to predict the yield performances of multicomponent oil smokes confined in closed chambers. The model employs vapor pressure-percent mass evaporated correlations, which were derived from oil droplet evaporation studies, to predict the saturation requirements of fog oil, PEG 200 and No. 2 diesel fuel. Comparison of model predictions and experiment indicate good agreement for fog oil and PEG 200 vaporization/condensation yield tests. Both model and (Continued on reverse side) | | |

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20. ABSTRACT (continued)

experiment demonstrate the inferior yield performance of No. 2 diesel fuel, possessing a yield less than half that of either fog oil or PEG 200. The lower yield performance of No. 2 diesel fuel is due to the high volatility of diesel fuel as compared to fog oil or PEG 200. The model also illustrates the dependency of multicomponent oil yield performances on chamber volume. As a result, while fog oil and PEG 200 will be characterized by similar yields at a specific chamber volume, the yield equivalency will not hold at larger chamber volumes. Implications on the liquid smoke-open atmosphere persistency characteristics are discussed. Finally, the hygroscopic nature of PEG 200 is discussed and a model is developed for the prediction of the yield performance of PEG 200 as a function of relative humidity. It is also shown the yield of PEG 200 can exceed that of fog oil under high-relative humidity conditions.

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PREFACE

The work described in this report was authorized under Project 1L161102A71A, CB Defense and General Investigations, Aerosol/Obscuration Science. This work was conducted from January to November 1980. The background data are contained in Notebook 9990.

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PREDICTING THE YIELD PERFORMANCES OF MULTICOMPONENT OIL SMOKES CONFINED IN CLOSED CHAMBERS

1. INTRODUCTION

Recently Vervier and Anderson (1978) measured the yield performances of various multicomponent oil smokes confined in closed chambers. The researchers defined the yield of a specific smoke as the ratio of the mass of smoke in a closed chamber to the mass of liquid used to generate the smoke. By mass filter sampling and assuming perfect aerosolization efficiency, the yield performances of fog oil SCF-2, PEG 200 and No. 2 diesel fuel were determined. The results indicated that fog oil and PEG 200 exhibited similar yield performances while No. 2 diesel fuel possessed a yield less than half that of the other oils.

In this paper a semiempirical model is developed to predict the yield performances of fog oil, PEG 200 and No. 2 diesel fuel smokes confined in closed chambers. The model is based on the oil droplet evaporation experiments of Rubel (1981). Rubel measured the evaporation rates of fog oil, PEG 200 and No. 2 diesel fuel droplets and derived vapor pressure-percent mass evaporated correlations for the oils. By employment of these vapor pressure correlations the saturation requirements of fog oil, PEG 200 and diesel fuel were quantified.

It is shown that model and experiment are in good agreement for the vaporization/condensation yield tests involving fog oil and PEG 200. Both model and experiment indicate a No. 2 diesel fuel yield performance which is less than half that of the yields of fog oil and PEG 200. The model also demonstrates the dependency of the smoke yields on the chamber volume. While Vervier and Anderson (1978) found the yields for fog oil and PEG 200 to be almost equal for the chamber volume of 190 m^3 , model calculations indicate fog oil yields exceed PEG 200 yields for larger chamber volumes. Consequently conclusions on the comparative persistency of these oil smokes based on yield performance measurements in closed chambers may be incorrect.

Finally the relative-humidity-dependent yield performance of hygroscopic PEG 200 is modelled from present vapor pressure-percent-mass-evaporated correlations. It is shown under high-relative-humidity conditions, the yield performance of PEG 200 can exceed that of fog oil.

2. MULTICOMPONENT OIL VAPOR PRESSURE CORRELATIONS

For a single-component oil the liquid vapor pressure is invariant with respect to evaporation. However, for multicomponent oils, the mixture vapor pressure becomes progressively lower as greater proportions of relatively nonvolatile species predominate in the liquid. One method for characterizing the composition-dependent vapor pressure of the mixture oils is through a distillation curve. But to apply this data to ambient conditions, an approximate extrapolation procedure is required which tends to weaken the validity of the results.

Rubel (1981)² measured the evaporation rates of multicomponent oil droplets which were suspended in an electrodynamic chamber (Frickel et al., 1979).³ By balancing the weight of the droplet against an opposing direct current voltage, successive droplet mass proportions were determined from successive voltage proportions. By recording the time histories of the droplet masses and applying continuum diffusion theory, the vapor pressure of the mixture droplet was determined as a function of percent mass evaporated.

Table 1 shows the vapor pressure correlations for fog oil, PEG 200 and No. 2 diesel fuel as derived from the droplet evaporation data (Rubel, 1981).

Table 1. Vapor Pressure Correlations for Fog Oil, PEG 200 and No. 2 Diesel Fuel at 25°C

| Liquid | Pressure correlations | Temperature °C | |
|-------------------|-------------------------------|-------------------|-----|
| Fog oil SGF-2 | $Lnp = 23.10 + 5.042 \ln m_r$ | 25 | (1) |
| PEG 200 | $Lnp = 10.71 + 2.467 \ln m_r$ | 25 | (2) |
| No. 2 diesel fuel | $Lnp = 14.39 + 4.423 \ln m_r$ | 25 | (3) |

The above relations were obtained by a least square linear regression analysis of the vapor pressure data and account for at least 98% of the data scatter. All units are in cgs units so pressure has the dimensions ergs/cc. The percent mass remaining m_r , defined as

$$m_r = 100 - m_e = (m/m_0) \times 100 \quad (4)$$

where

m_0 is the initial droplet mass, m is the instantaneous mass and m_e is the percent mass evaporated, ranging as $0 \leq m_r \leq 100$. The liquids under study were obtained by vaporizing the smoke liquid and recondensing through a water-cooled jacket. The recondensed liquids were then suspended in the electrodynamic chamber for analysis.

For No. 2 diesel fuel the droplet evaporation rate is several orders of magnitude greater than that of fog oil or PEG 200. As a result the determination of the initial droplet mass was difficult by the electrodynamic balance method. To determine the percent mass remaining at a given time, the distillation curve for No. 2 diesel fuel was employed. From the distillation curve the vapor pressure at a given mass remaining is extrapolated to ambient temperatures using the Clausius Clapeyron relation. Matching this vapor pressure with vapor pressure value derived from droplet evaporation data, the initial droplet mass is determined.

3. ALGORITHM FOR PREDICTING SMOKE YIELD PERFORMANCE

The yield performance (YP) of an arbitrary nonreacting, nonabsorbing smoke is

$$YP = 1 - m_v/m_0 \quad (5)$$

where

m_v is the mass of vapor, given up by the smoke, and

m_0 is the initial mass of liquid used to generate the smoke. For a single-component liquid smoke the yield is

$$YP = 1 - M_w p(T) V / RT m_0 \quad (6)$$

where

V is the chamber volume,

M_w the liquid molecular weight, and $p(T)$ is the equilibrium vapor pressure of the liquid at the temperature T .*

Applying Dalton's law, the mass associated with a multicomponent vapor is

$$m_v = V \sum_i p_i(T) M_{wi} / RT \quad (7)$$

and the yield performance is

$$YP = 1 - V \sum_i p_i(T) M_{wi} / RT m_o \quad (8)$$

where

$p_i(T)$ and M_{wi} are the partial pressure and molecular weight of the i th species respectively.

Experimental determination of $p_i(T)$ is difficult and theoretical modelling of the partial pressure as a function of percent evaporated requires, as of yet unavailable data on the initial-species solution concentration. Consequently the prediction of smoke yields through equation 6 is not practical.

To bypass the aforementioned problem the following algorithm is presented for the prediction of the yield performance of multicomponent oil smokes. Chamber saturation is achieved once the liquid vapor pressure equals the environmental vapor pressure. Actually equilibrium is achieved once the environmental saturation ratio is equal to the droplet saturation ratio. However, since both aerosol and chamber are in thermal equilibrium then equality of saturation ratios is equivalent to equality of pressures. The liquid vapor pressure is a function of percent mass remaining and is given by equations 1 through 3. The environmental vapor pressure is given as the sum of partial pressures

$$p_e = RT \sum_i m_{vi} / M_{wi} \quad (9)$$

If the species molecular weight is assumed constant then equation 9 becomes

$$p_e = RT m_v / M_w \quad (10)$$

As is shown in table 2 for the range of species molecular weights characteristic of the oil mixtures, the variation in molecular weight results in a small variation in the calculated yield performance. For example, the fog oil species molecular weight varies from 250 gm/mole to 400 gm/mole (Baek, et al., 1980)⁴ while the calculated yield varies only from 0.93 to 0.95. Diesel fuel and PEG 200 are a narrower mixture of oil species and variations in species molecular weight will have an even smaller effect on the calculated yields than in the case of fog oil.

The mass of vapor m_v evaporated from the smoke can be derived from a mass balance and can be expressed as

$$m_v = m_o (1 - m_r/100) \quad (11)$$

* In the present analysis the vapor-pressure-elevation Kelvin effect is neglected due to the large aerosol sizes studied by Vervier and Anderson.¹

Table 2. Yield Performance of Multicomponent Oil Smokes Determined Experimentally and Calculated from Equation 12

| Liquid | Dissemination | Vervier-Anderson | Model |
|--------------|---------------|------------------|--------|
| Fog oil SGF2 | V/C* | 0.93 | 0.94** |
| Fog oil SGF2 | Pneumatic | 0.75 | 0.92 |
| PEG 200 | V/C | 0.89 | 0.92 |
| PEG 200 | Pneumatic | 0.77 | 0.90 |
| Diesel | V/C | 0.19 | 0.40 |
| Diesel | Pneumatic | 0.48 | 0.49 |

* The dissemination method V/C is vaporization/condensation.

** The yield 0.94 is an average of 0.93, 0.94 and 0.95, the variation in yield due to the variation in species molecular weight, i.e., $250 \text{ gm/mole} \leq M_{wi} \leq 450 \text{ gm/mole}$.

Equating the environmental vapor pressure p_e , equation 10, and the liquid vapor pressure p_l , equations 1 through 3, the condition for saturation becomes

$$p_l(m_r) = m_o RT(1 - m_r/100)/(M_{wi}V) \quad (12)$$

Equation 12 is a transcendental equation in the mass remaining m_r and is solved numerically. Table 2 shows the yield performances of fog oil, PEG 200 and No. 2 diesel fuel as calculated from equation 12 and experimentally determined by Vervier and Anderson (1979).¹

Table 2 shows that there exists good agreement between the semiempirical model and the data of Vervier and Anderson (1979)¹ for the fog oil and PEG 200 vaporization/condensation yield tests. Model yields slightly overestimate experimental yields for the pneumatic dissemination yield tests of fog oil and PEG 200. This yield disparity is probably due to the less than 100% pneumatic aerosolization efficiency.

Both model and experiment indicate the yield of diesel fuel is substantially less than that of fog oil or PEG 200. While some disparity exists between model and experiment for the vaporization/condensation tests, excellent agreement is found for the pneumatic aerosolization yield test. Possibly the vaporization process causes a thermal decomposition of the diesel vapor. The thermal cracking can lead to an elevation in vapor pressure and a concomitant reduction in yield. The elevation in mixture vapor pressure has been experimentally evidenced for fog oil by Rubel (1981)² and Baek et al. (1980).⁴ The data in table 2 for fog oil and PEG 200 are derived from the evaporation rates of recondensed smokes thereby accounting for possible thermodynamic changes as a result of sudden vaporization. However distillation curves for recondensed diesel fuel do not exist and proper accounting for possible changes in mixture properties could not be achieved.

4. SMOKE YIELD DEPENDENCY ON CHAMBER VOLUME

It would seem reasonable to conclude from table 2 that the yield performance of fog oil and PEG 200 are approximately equal. However, the yields of the oils are a function of chamber volume. To clearly see this dependency, the yield performances of PEG and fog oil are calculated as a function of chamber volume in table 3.

Table 3. Chamber Volume Dependency of the Yield Performances of Multicomponent Oil Smokes

| Chamber volume 10^6 cm^3 | Yields | |
|---------------------------------------|---------|---------|
| | Fog oil | PEG 200 |
| 190 | 0.94 | 0.92 |
| 300 | 0.91 | 0.88 |
| 500 | 0.88 | 0.82 |
| 5000 | 0.68 | 0.50 |

The initial mass of liquid m_0 is held constant while the chamber volume is increased. Clearly as the chamber volume is increased, the yield of PEG 200 and fog oil become significantly different. The reason for the increasing disparity is that as the chamber volume is increased a greater proportion of liquid is evaporated in order to saturate the chamber. Since PEG 200 has a greater vapor pressure than fog oil for degrees of evaporation exceeding 10% (see equation 1), then the yield of PEG will be less than fog oil when the chamber volume requires liquid evaporation beyond 10%. Therefore extensions of comparative yield performances in closed chambers to cloud lifetimes in open atmospheres can be incorrect.

5. HYGROSCOPIC OIL SMOKE YIELD DEPENDING ON RELATIVE HUMIDITY

The yield performance algorithm presented in section 3 applied to nonreacting, nonabsorbing oil smokes. While fog oil and No. 2 diesel fuel are nonhygroscopic hydrocarbon mixtures, PEG 200 is hygroscopic and will absorb increasing quantities of water as the relative humidity is increased.

The hygroscopic nature of PEG 200 alters the smoke yield performance in two ways. First the absorption of water by a PEG 200 mixture reduces the vapor pressure of the oil mixture in accordance with thermodynamic considerations. Thus, by reducing the aerosol oil vapor pressure, a smaller quantity of oil vapor is required to saturate the closed chamber and the resultant yield will be increased. Secondly the addition of water (beyond which is originally in the oil) represents the addition of mass to the aerosol and also results in an increased smoke yield. Allowing for the presence of water in the original PEG 200 bulk the yield performance is

$$YP = \frac{m^a}{m^b} = \frac{m^a_{\text{oil}} + m^a_{\text{H}_2\text{O}}}{m^b_{\text{oil}} + m^b_{\text{H}_2\text{O}}} = \frac{m^a_{\text{oil}}}{m^b_{\text{oil}}} \left[\frac{1 + m^a_{\text{H}_2\text{O}}/m^a_{\text{oil}}}{1 + m^b_{\text{H}_2\text{O}}/m^b_{\text{oil}}} \right] \quad (13)$$

where a and b are aerosol and bulk states respectively.

From droplet evaporation data, PEG 200 oil mixture vapor pressure was defined in terms of the mass remaining parameter m_r (equation 2). Since the oil vapor pressure is a function of quantity of water absorbed, then the particular correlation 2 is valid at a particular relative humidity only, in the present case RH = 50%.

The percent mass remaining in the droplet is given by

$$m_r = \left[\frac{m_{oil}^f + m_{H_2O}^f}{m_{oil}^i + m_{H_2O}^i} \right] \times 100 = \left[\frac{m_{oil}^f}{m_{oil}^i} \right] \left[\frac{1 + m_{H_2O}^f/m_{oil}^f}{1 + m_{H_2O}^i/m_{oil}^i} \right] \times 100 \quad (14)$$

Now the volatility of water is much greater than that of any species of PEG 200 so the water mass transfer rate is much greater than that of PEG 200. During the droplet evaporation of PEG 200, the water species will then be in thermodynamic equilibrium with respect to the rate of oil mass transfer. The mass ratio m_{H_2O}/m_{oil} is the solution concentration and the chemical potential in the gas phase which establishes equilibrium between the water chemical potential in the liquid phase μ_{H_2O} . The water chemical potential in the gas phase is

$$\mu_{H_2O} = \mu_{H_2O}^0 + RT \ln (p_{H_2O}/p_{H_2O}^0) \text{ (gas)} \quad (15)$$

where

μ_i^0 is the standard chemical potential at p_i^0 and we assume the gas is ideal. The fact the specific free energy (μ) is defined relative to some standard state, $\mu_{i,0}$ is the reason the thermodynamic properties of solutions depend on the relative humidity, p_i/p_i^0 as opposed to the absolute humidity p_i .

The water chemical potential in the liquid phase is

$$\mu_{H_2O} = \mu_{H_2O}^0 + RT \ln a_{H_2O} \quad \text{(liquid)} \quad (16)$$

At equilibrium

$$(\mu_{H_2O})^l = (\mu_{H_2O})^g \quad \text{and}$$

$$RH = a_{H_2O} \quad (17)$$

where

we use the definition $p_{H_2O}/p_{H_2O}^0 = RH$.

The activity is defined in terms of the activity coefficient α and the mole fraction X as

$$a_{H_2O} = \alpha(X_{H_2O}) X_{H_2O} \quad (18)$$

and is thus a function of composition mole fraction X_{H_2O} . Then at constant relative humidity, the mole fraction composition is invariant. Since the mass fraction is determined by the mole fraction then at constant relative humidity the mass fraction is invariant too and

$$m_{H_2O}^f/m_{oil}^f = m_{H_2O}^i/m_{oil}^i \quad (19)$$

The equality is approximate since it is assumed the activity of water is independent of the particular PEG 200 molecule comprising the solution. This assumption can be experimentally verified by measuring the growth rates of PEG 200 droplets at different stages of the droplet evaporation. The activity of water is thus determined in different compositions of PEG 200 molecules.

Figure 1 shows the growth with relative humidity of a PEG 200 droplet measured in an electrodynamic suspension chamber after negligible oil evaporation. Future research will be directed toward measuring the growth rates of PEG 200 molecules at different compositions. However, for the present closed-chamber studies, where yields are about 0.90, the growth data of the droplets after negligible evaporation should be a good approximation of the oil thermodynamics.

Returning to equation 14 and using equation 19 gives

$$m_r = m_{oil}^f / m_{oil}^i \times 100$$

With $m_{oil}^f / m_{oil}^i = m_{oil}^a / m_{oil}^b$, the yield performance becomes

$$YP = \frac{m_r}{100} \left[\frac{1 + m_{H_2O}^a / m_{oil}^a}{1 + m_{H_2O}^b / m_{oil}^b} \right] \quad (20)$$

Both m_r and m_{H_2O} / m_{oil} are functions of relative humidity. The growth data in figure 1 quantifies $m_{H_2O}^a / m_{oil}^a$ and $m_{H_2O}^b / m_{oil}^b$ where it is envisioned that the bulk sample of PEG 200 has been allowed to come to equilibrium with some storage relative humidity. Or perhaps the bulk relative humidity corresponds to the relative humidity at processing; clearly the bulk solution is not a well-defined quantity and a sensitivity analysis will be conducted to ascertain the dependence of yield performance on initial bulk composition.

The dependence of m_r on relative humidity is determined as follows. The vapor pressure-percent mass remaining correlation for PEG 200 is given by equation 2 for a relative humidity of 50%. For a different relative humidity, a different amount of water is present in solution and the oil vapor pressure is altered.

With

$$p_{oil} = p_{oil}^o a_{oil} \quad (21)$$

where p_{oil}^o is the mixture vapor pressure in the presence of no water, then the mixture pressure at different relative humidities is

$$\frac{p_{oil}(RH_2)}{p_{oil}(RH_1)} = \frac{a_{oil}(RH_2)}{a_{oil}(RH_1)} \quad (22)$$

assuming an ideal solution, then $a_{oil} = x_{oil} = 1 - x_{H_2O}$ and

$$p_{oil}(RH) = p_{oil}(50\%) \left(\frac{x_{oil}(RH)}{x_{oil}(50\%)} \right)$$

or

$$p_{oil}(RH) = p \left(\frac{x_{oil}(RH)}{x_{oil}(50\%)} \right)$$

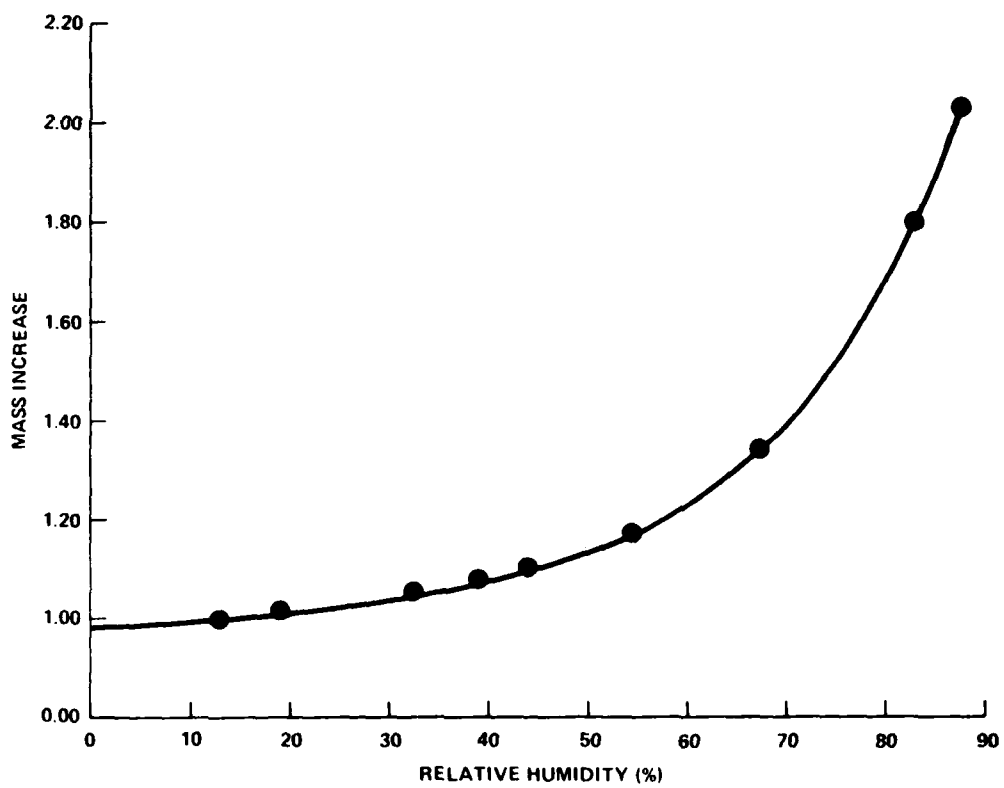


Figure 1. Growth Rate of PEG 200 Droplet Measured
in an Electrodynamic Suspension Chamber

where p is the reported value in equations 1 through 3. The oil activity a_{oil} could be derived from the activity of water using the Gibbs Duhem relation. However, it is expected the ideal solution approximation is not severe.

Using equation 2

$$\ln p(RH) = -10.71 + \ln \left[\frac{x_{oil}(RH)}{x_{oil}(50\%)} \right] + 2.4671 n m_r \quad (23)$$

Using equation 12 gives

$$e^{-10.71} \frac{x_{oil}(RH)}{x_{oil}(50\%)} m_r^{2.467} = \frac{m_o RT (1 - m_r/100)}{V M_w} \quad (24)$$

Thus the mass remaining is determined by first specifying the relative humidity. From the growth curve, figure 1, the oil mole fraction is determined. The transcendental equation 24 is solved numerically and the value of m_r is substituted into equation 20 for calculation of the yield performance. The equilibrium mass ratio $m^a_{H_2O}/m^a_{oil}$ is also determined by the relative humidity. The mass ratio $m^b_{H_2O}/m^b_{oil}$ is postulated to be the water mass ratio at the storage relative humidity.

Figure 2 shows the yield performances of PEG 200 as a function of relative humidity and parameterized with respect to the storage relative humidity (SR). In all cases the yield performance is a monotonically increasing function of relative humidity. As the storage relative humidity increases, the yield performance decreases. This results from the increasing amounts of water present in the bulk oil negating the effect of water addition in the aerosol state. Also shown in figure 2, is the intersection between the yield performance given by Vervier and Anderson (1979)¹ of 0.89 and the isopleths of storage relative humidity. Thus the yield performance of PEG 200 at an environmental relative humidity of 45% and with an initial storage relative humidity of around 50% would give a yield performance of 0.89.

Such a storage relative humidity would correspond to an oil mass fraction of 87% at storage. It must be emphasized that it is assumed perfect aerosolization is achieved in the present calculations. So agreement with experiment can be achieved even at lower storage relative humidities by introducing aerosolization efficiency corrections. Finally due to water absorption by PEG 200, the yield performance of this oil can exceed that of fog oil at high relative humidities, in the present case for relative humidities exceeding 50% and storage relative humidities less than 30%.

6. CONCLUSIONS

1. A semiempirical model is developed to predict the yield performances of multi-component oil smokes which are confined in closed chambers.

2. The model yield values agree quantitatively with experimental data indicating the yield performance of No. 2 diesel fuel is less than half that of PEG 200 or fog oil under the conditions studied.

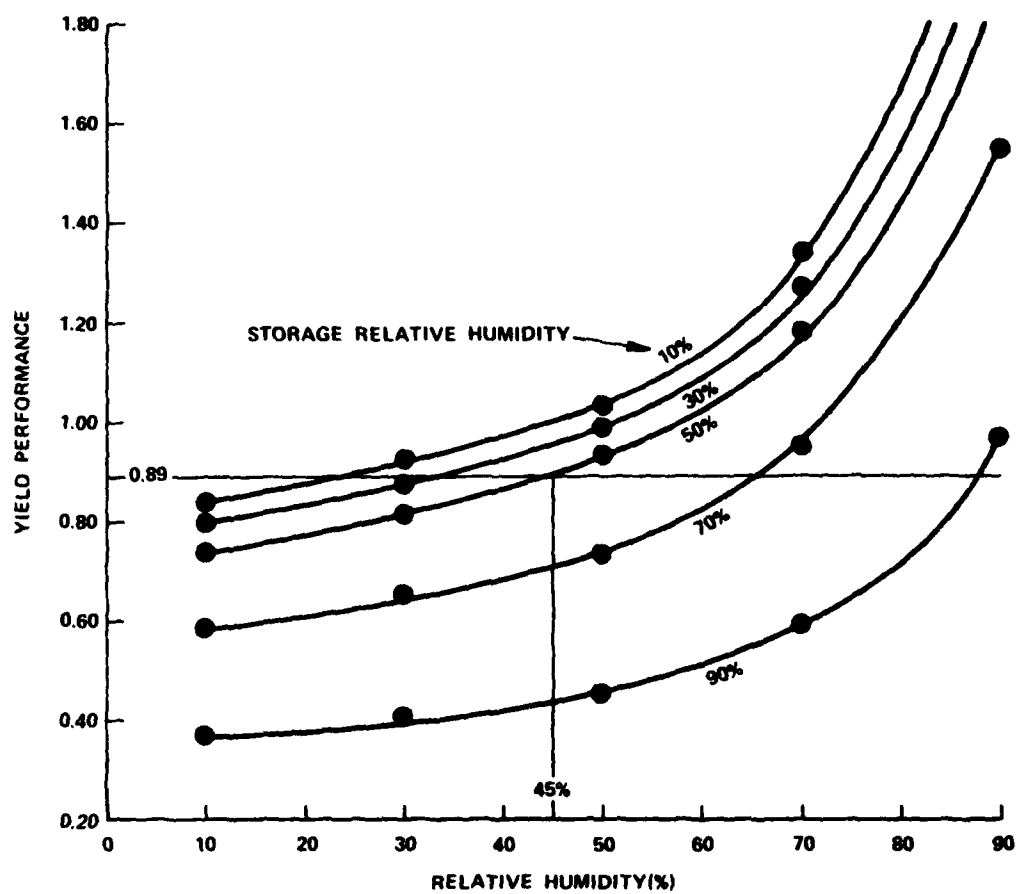


Figure 2. Relative-Humidity-Dependent Yield Performance of PEG 200

3. The closed-chamber yield performances are chamber volume dependent and while PEG 200 and fog oil exhibit similar yields at the chamber volume of 190 m^3 at larger volumes the yield performance of fog oil will exceed that of PEG 200.

4. Due to the hygroscopic nature of PEG 200, the yield performance of PEG 200 can exceed that of fog oil at high relative humidities.

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GLOSSARY

| | |
|----------------|--|
| m_e | - Percent mass evaporated |
| m_r | - Percent mass remaining |
| m_v | - Mass concentration of vapor (gram/cm ³) |
| M_w | - Molecular weight (gram/mole) |
| M_{wi} | - Molecular weight of ith species (grams/mole) |
| p_e | - Total environmental vapor pressure (dyne/cm ²) |
| p_i | - Vapor pressure of ith species (dyne/cm ²) |
| p_l | - Liquid vapor pressure (dyne/cm ²) |
| R | - Gas constant (ergs/mole-°K) |
| V | - Chamber volume (cm ³) |
| T | - Temperature |
| m^a | - Mass in aerosol state (gm/cm ³) |
| m^b | - Mass in bulk state (gm/cm ³) |
| m^f | - Final mass of droplet (gm) |
| m^i | - Initial mass of droplet (gm) |
| μ_{H_2O} | - Chemical potential of water (ergs) |
| $\mu^0_{H_2O}$ | - Standard chemical potential of water (ergs) |
| p_{H_2O} | - Partial pressure of water (ergs/cm ³) |
| $p^0_{H_2O}$ | - Equilibrium vapor pressure of water over plane surface of pure water (ergs/cm ³) |
| a_{H_2O} | - Activity of water |
| RH | - Relative humidity |
| SR | - Storage relative humidity |
| α | - Activity coefficient |
| x | - Mole fraction |

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